Nanoscale lattice dynamics probed by extreme ultraviolet transient gratings

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Outline

1) The transient grating (TG) approach

2) FEL-based TG, from demonstration to user operation

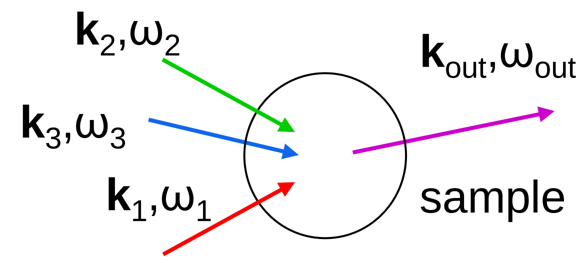
3) Nanoscale phonon dynamics and thermal transport

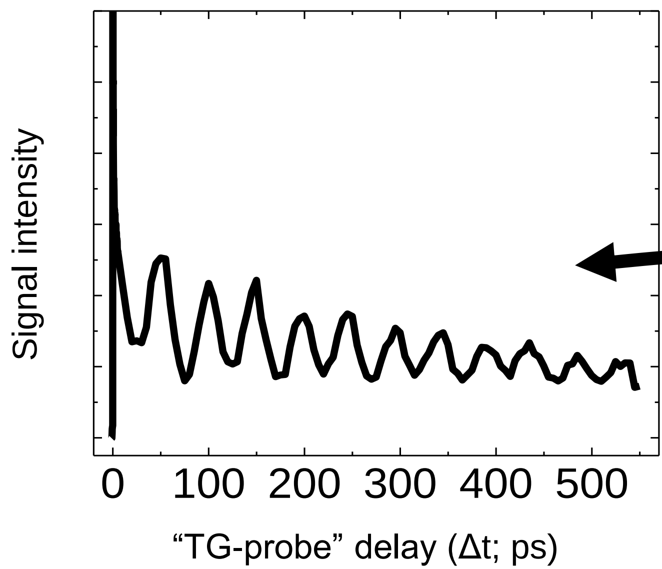
4) Beyond phonon and thermal dynamics

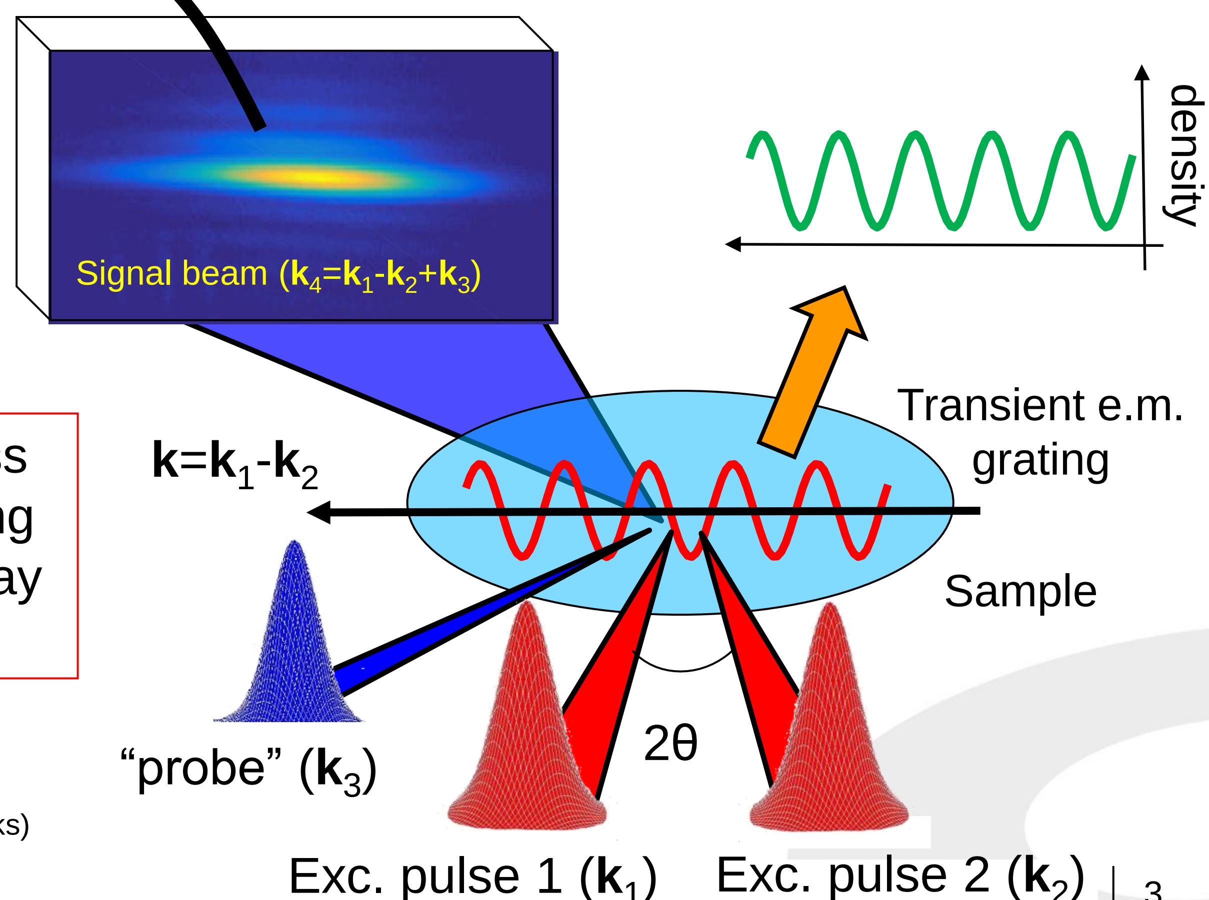
5) Conclusions

The transient grating (TG) method

$$P_i = \chi^{(1)}_{ij} E_i + \chi^{(2)}_{ijk} E_j E_k + \chi^{(3)}_{ijkl} E_j E_k E_l + \cdots$$



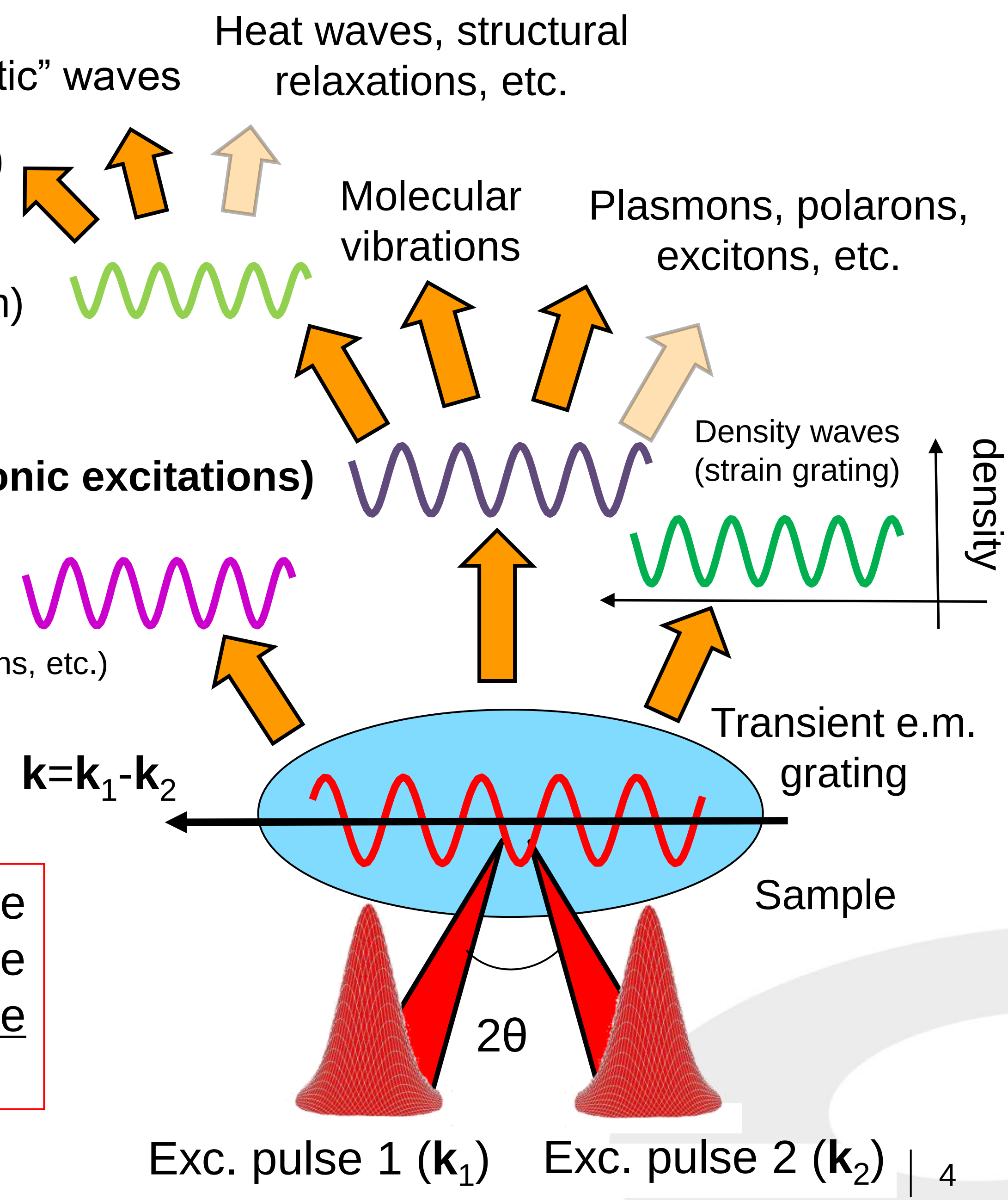




TG is a 3rd order non-linear process (four-wave-mixing, FWM) → starting point to develop other XUV/soft x-ray FWM experiments[1]

1) S. Tanaka and S. Mukamel, PRL 2002 (and many other works)

The TG method: an approach for probing different dynamics



$$L_{TG} = 2\pi/|\mathbf{k}| = \lambda_{FEL}/2\sin(\theta)$$

TG is a general approach, able to excite any dynamical variable coupled to the input fields, and <u>all these excitations are associated to a selected wavevector</u> (**k**)

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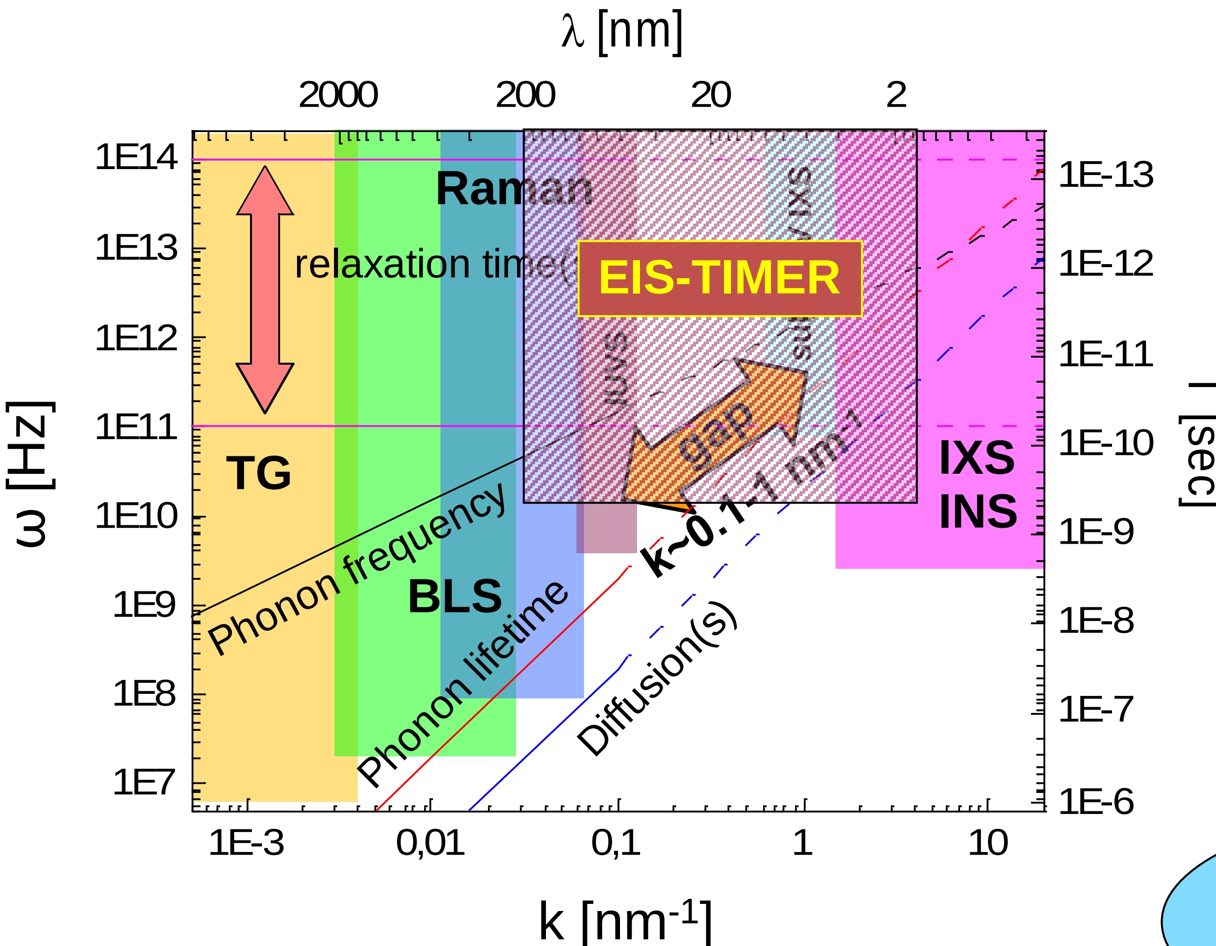
FEL-based EUV/soft x-ray TG (XTG)

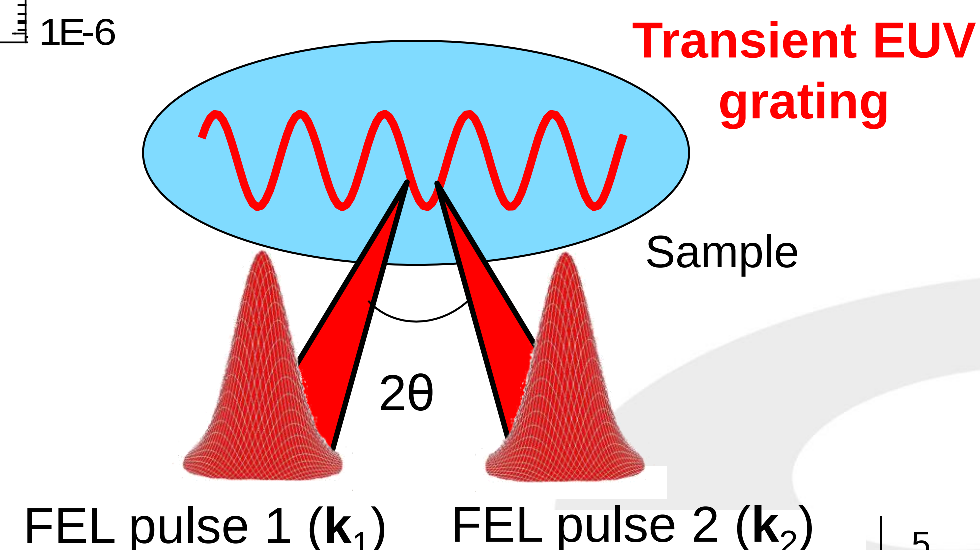


FERMI: λ_{FEL} = 60 – 4 nm ;
δt_{FEL} ~ 40 fs

EIS-TIMER BL: Δt_{max} ~ ns ;
$\theta = 9.2^o - 52.7^o$

$|\mathbf{k}|$ = 0.03 – 2 nm^{-1}

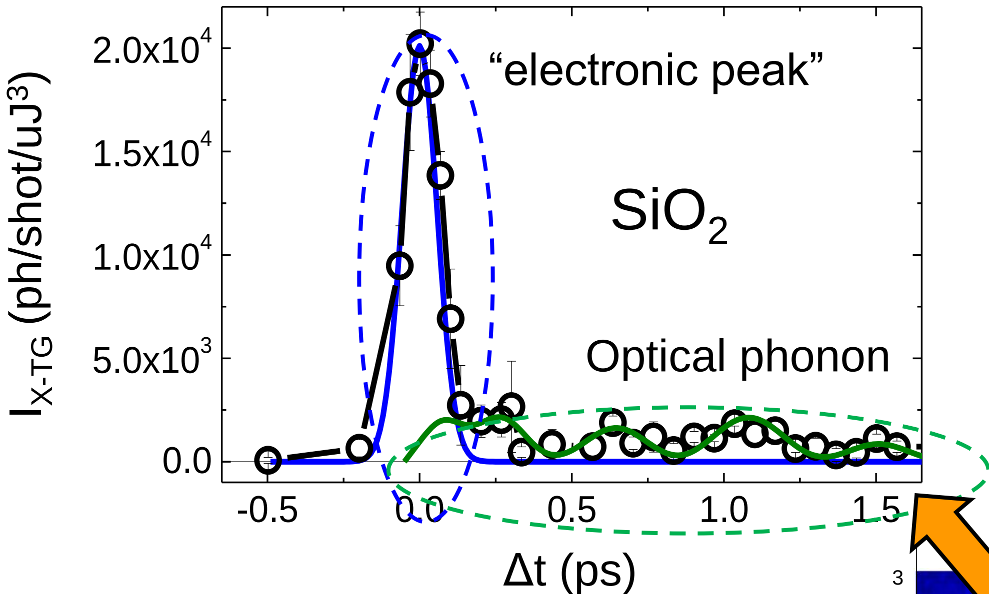
ω = 0.01 – 20 THz



Our initial aim was probing **collective lattice dynamics** (phonons, transport phenomena, structural relaxations, etc.) at **"mesoscopic" scales** (10's nm)

F. Bencivenga and C. Masciovecchio (NIMA 2009)

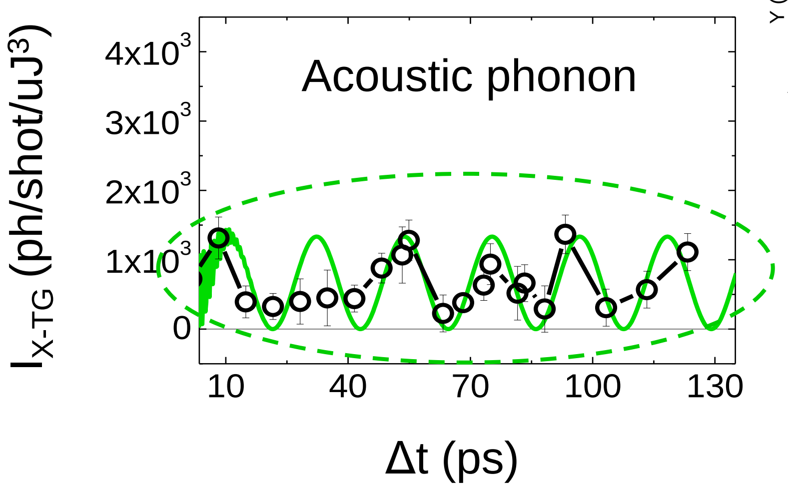
Firt signal !



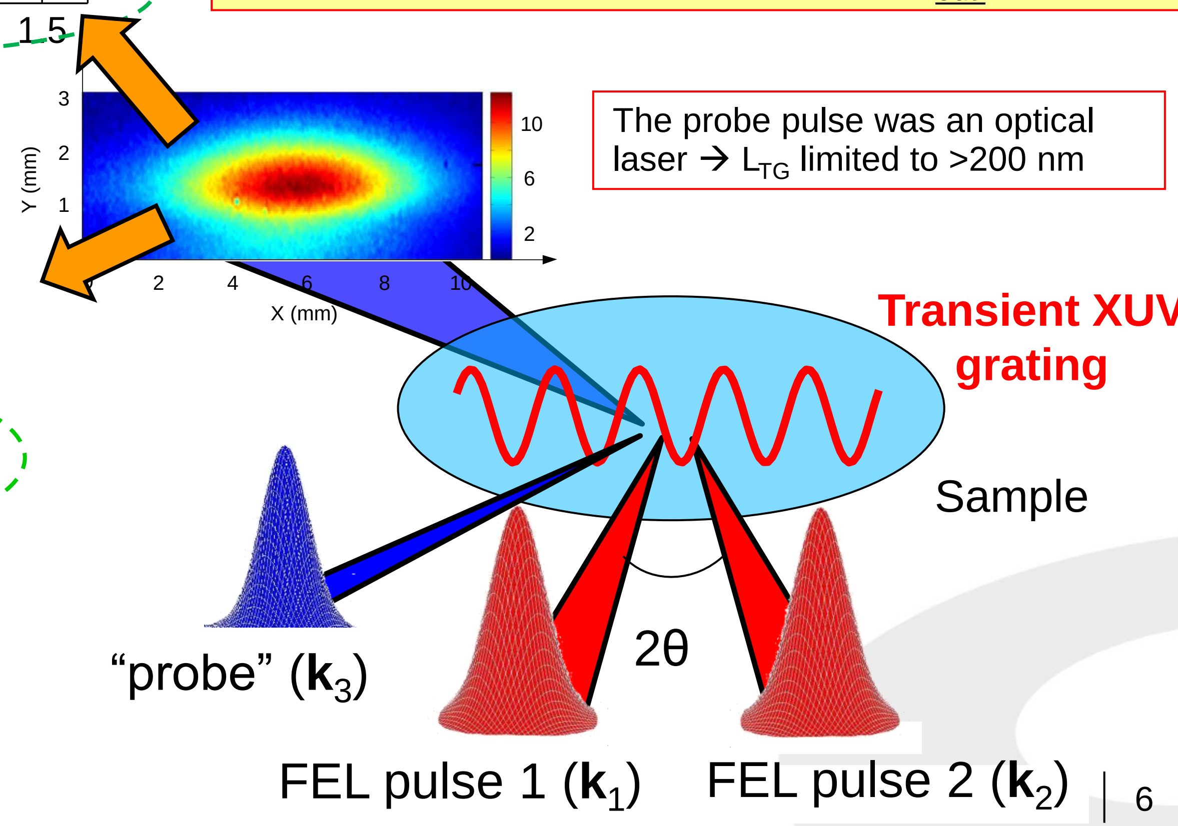
This works because we can generate XTG's on a 'macroscopic scale' (i.e. 10-100 μm), this needs **longitudinal coherence** (FERMI is a **seeed FEL**) of the excitation pulses: signal ~ $(L_{coh} / L_{TG})^2$

...experimental data were not super clear...

A "well defined" beam appears after time-zero downstream the sample along k_{out} → **FWM**



The probe pulse was an optical laser → L_{TG} limited to >200 nm



F. Bencivenga et al., Nature 2015

XTG two years later...

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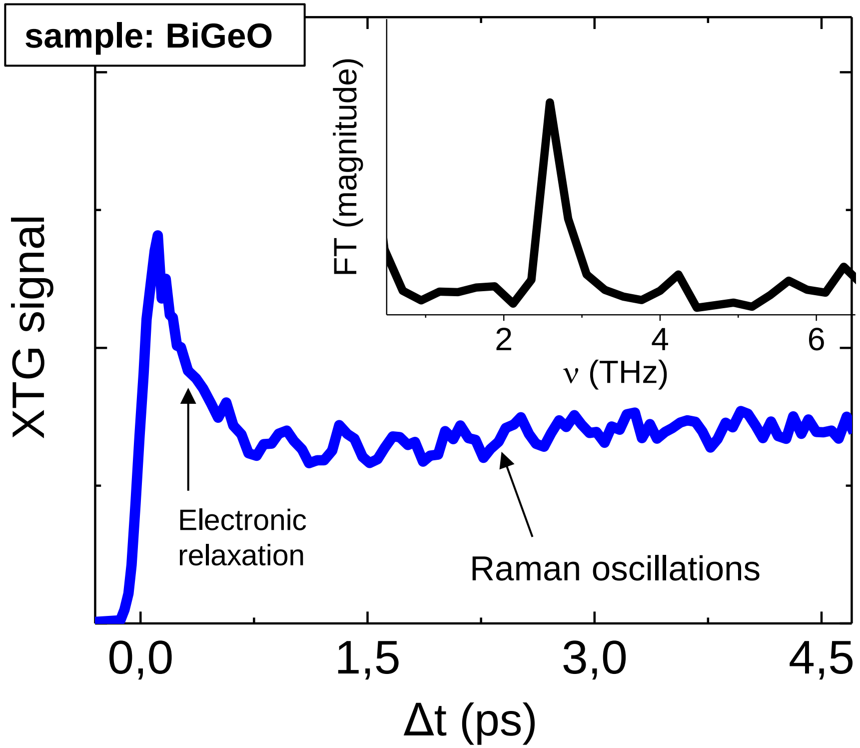
Molecular vibrations (stimulated Raman)



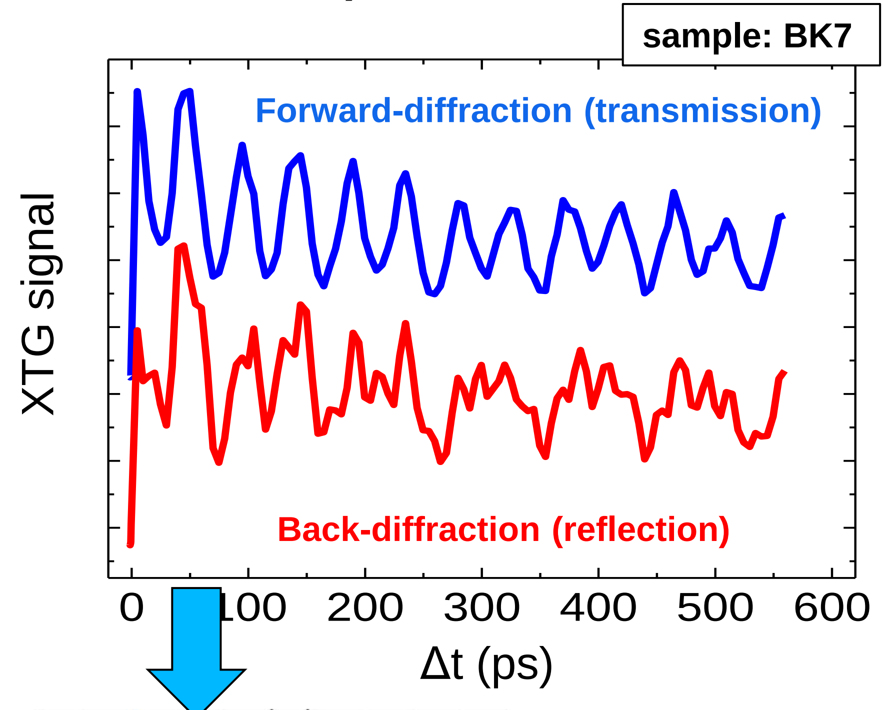
ω_{FEL}~100 eV

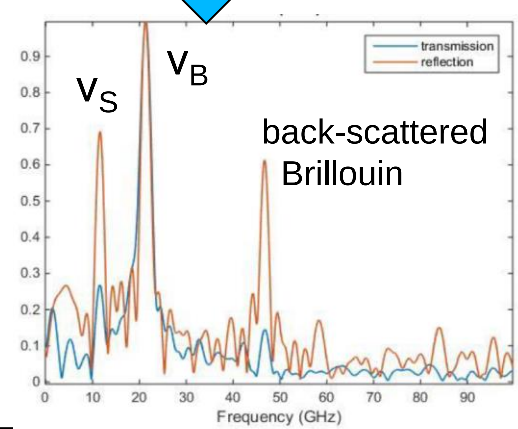
ω_{probe}~3 eV → L_{TG} ~ 270 nm

On longer timescales (not shown): clear phonon oscillations (10's GHz) and thermal decay (ns)

A. A. Maznev et al., APL 2018

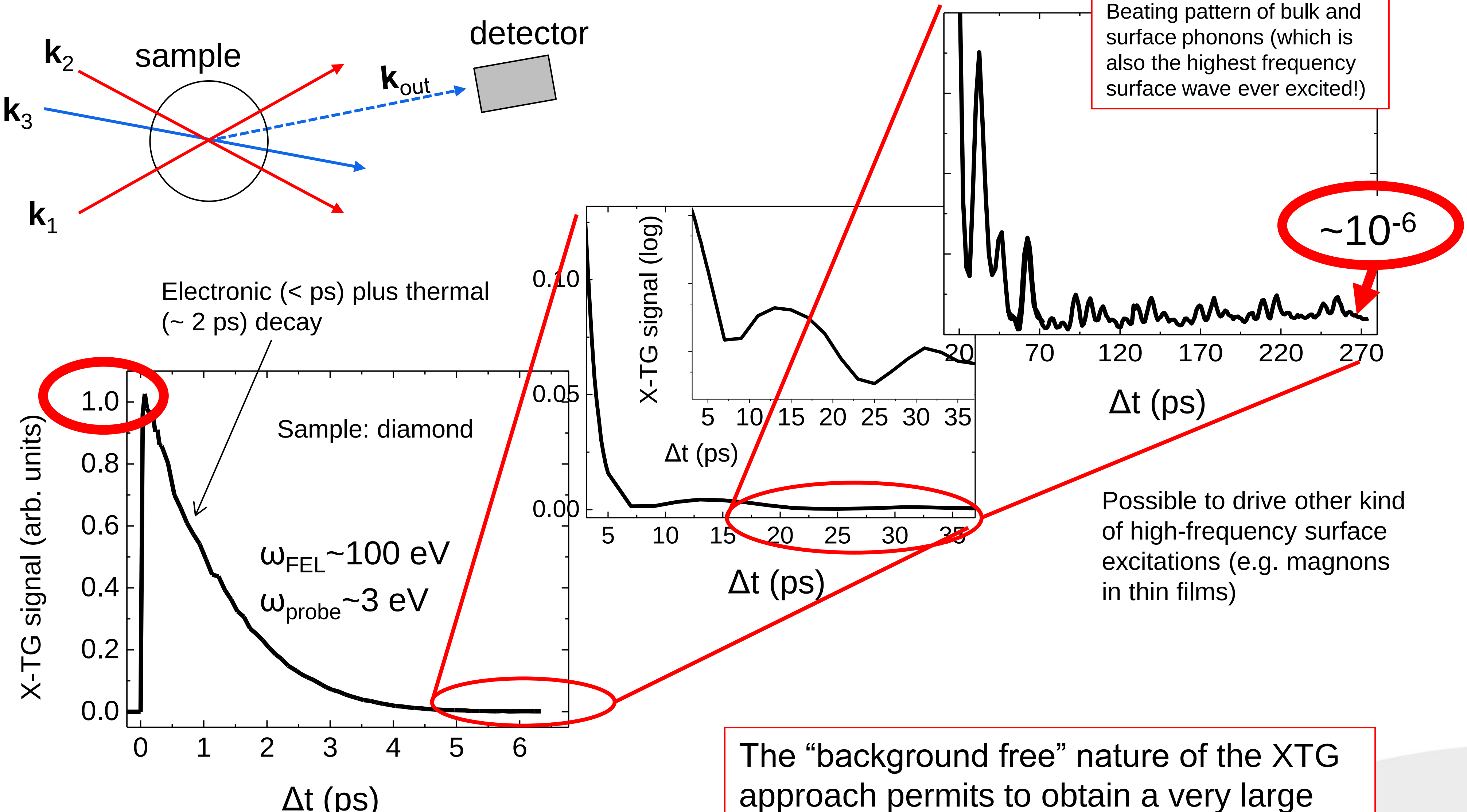
Bulk and surface phonons (forward vs back diffraction)





Quantitative analysis of surface and bulk contributions

XTG two years later...

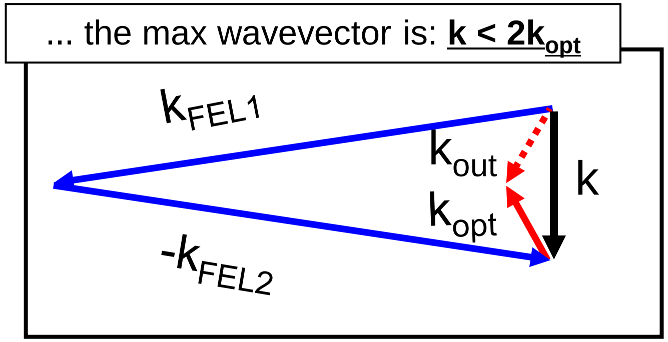


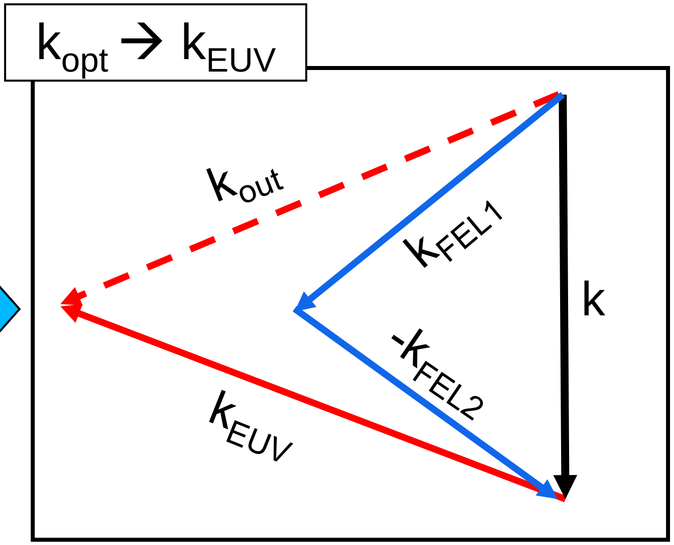
The “background free” nature of the XTG approach permits to obtain a very large contrast, compared to a “standard” FEL experiment

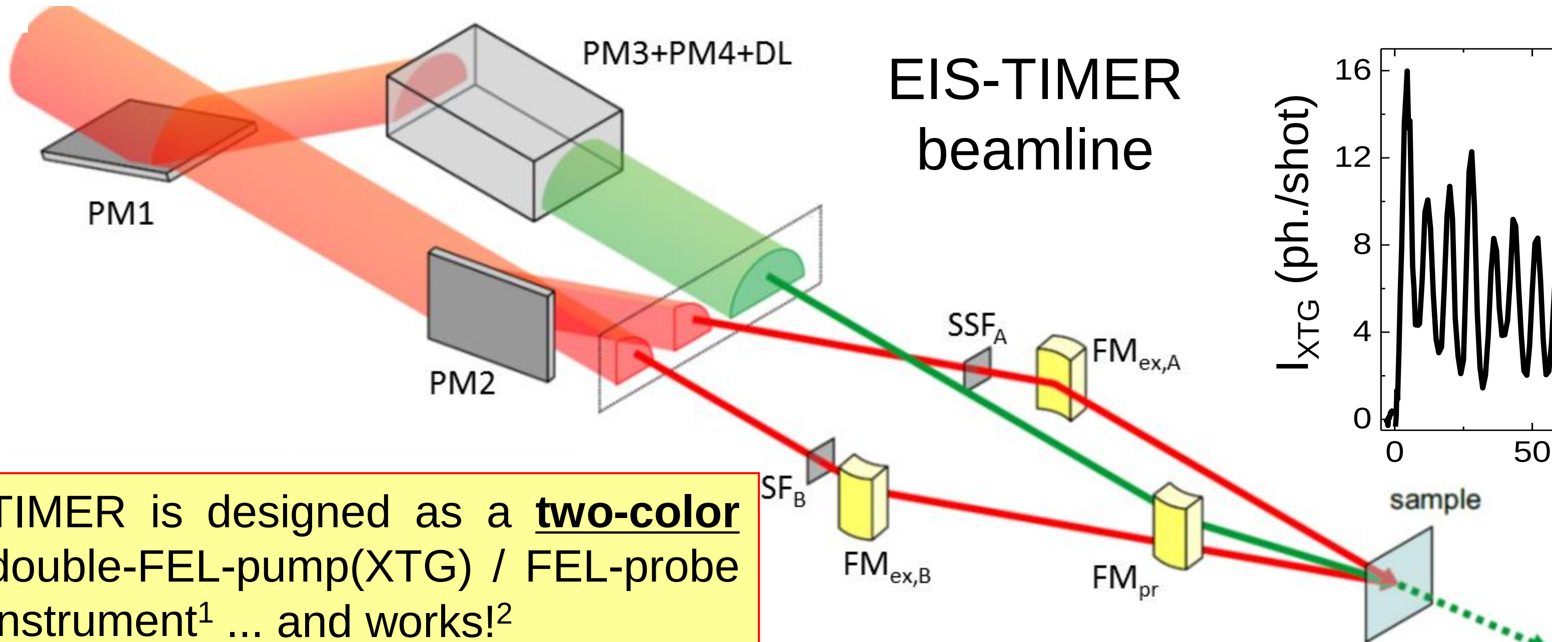
A. A. Maznev et al., APL 2018

XTG at the nanoscale

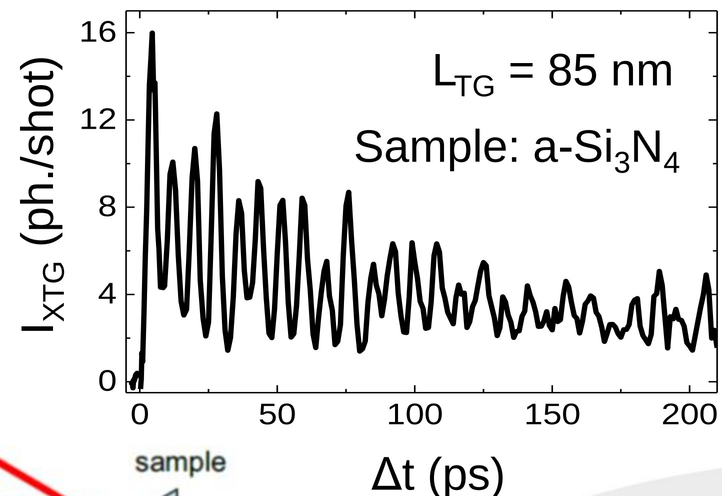
All I have shown up to now was obtained with optical probing, then...







EIS-TIMER beamline



TIMER is designed as a **two-color** double-FEL-pump(XTG) / FEL-probe instrument[1] ... and works![2]

1) R. Mincigrucci et al., NIMA (2018)

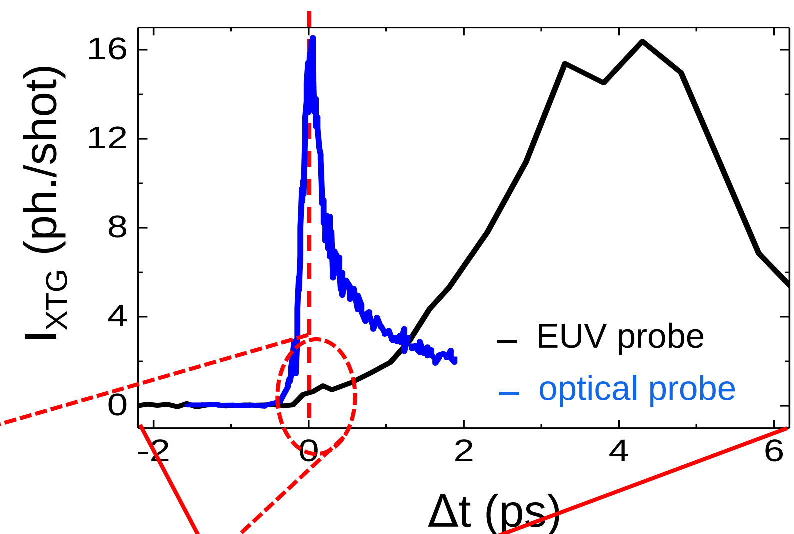
2) F. Bencivenga et al., Science Adv. (in press)

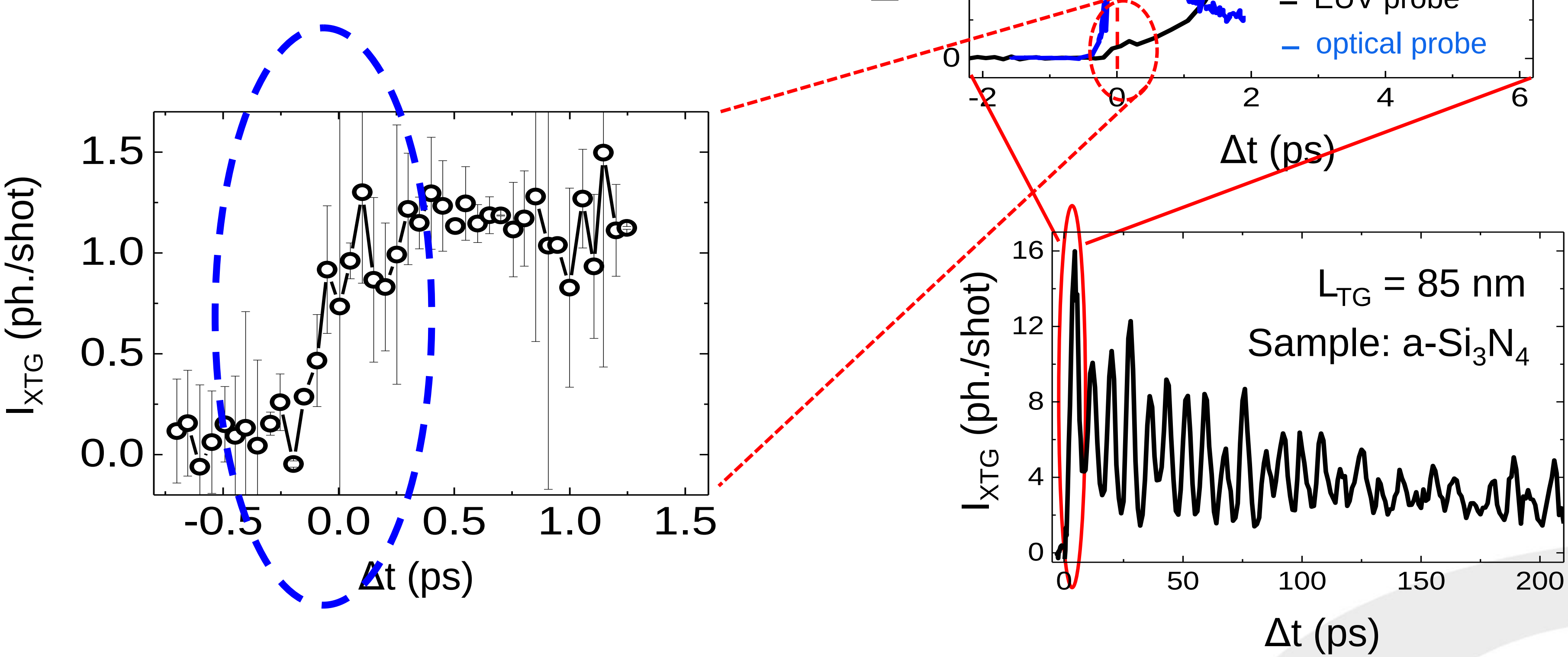
XTG at the nanoscale
- Optical vs EUV probing

The electronic response (electronic population grating) has a marginal contribution in EUV probed XTG.

→ EUV pulses are an excellent probe for lattice dynamics

→ EUV core-hole resonances needed to exploit the electronic (all-EUV FWM) response





F. Bencivenga et al., Science Adv. (in press)

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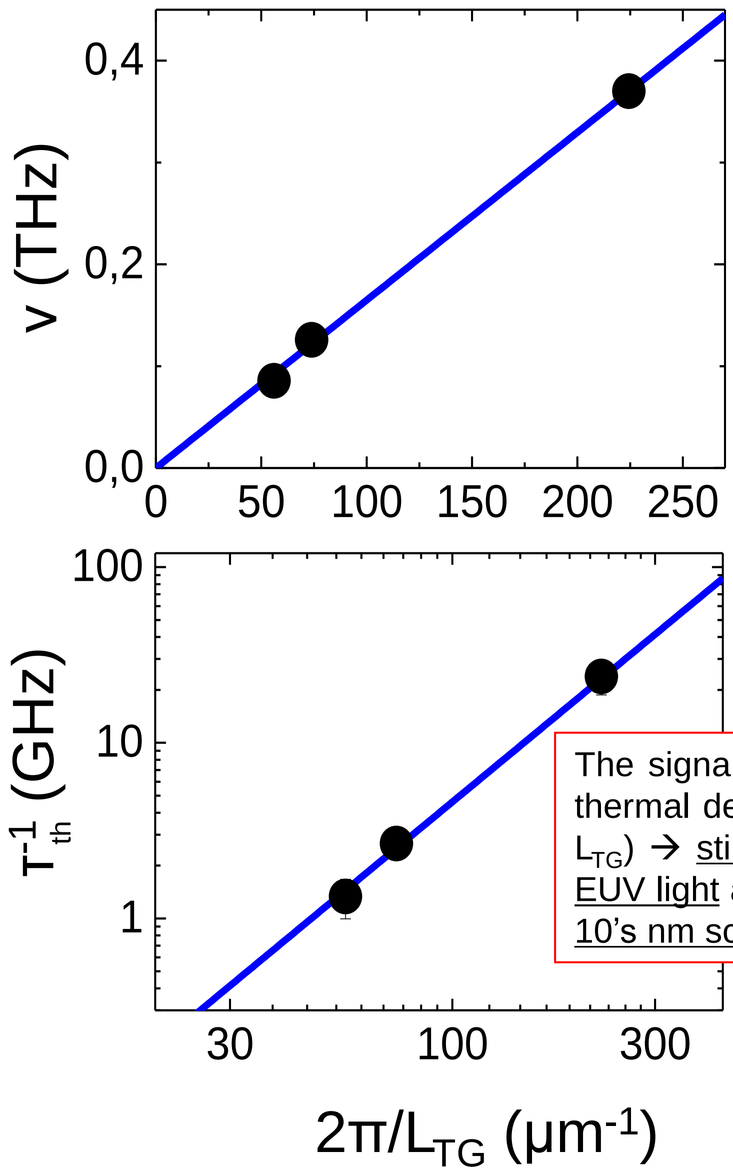
XTG at the nanoscale
- phonon and thermal dynamics

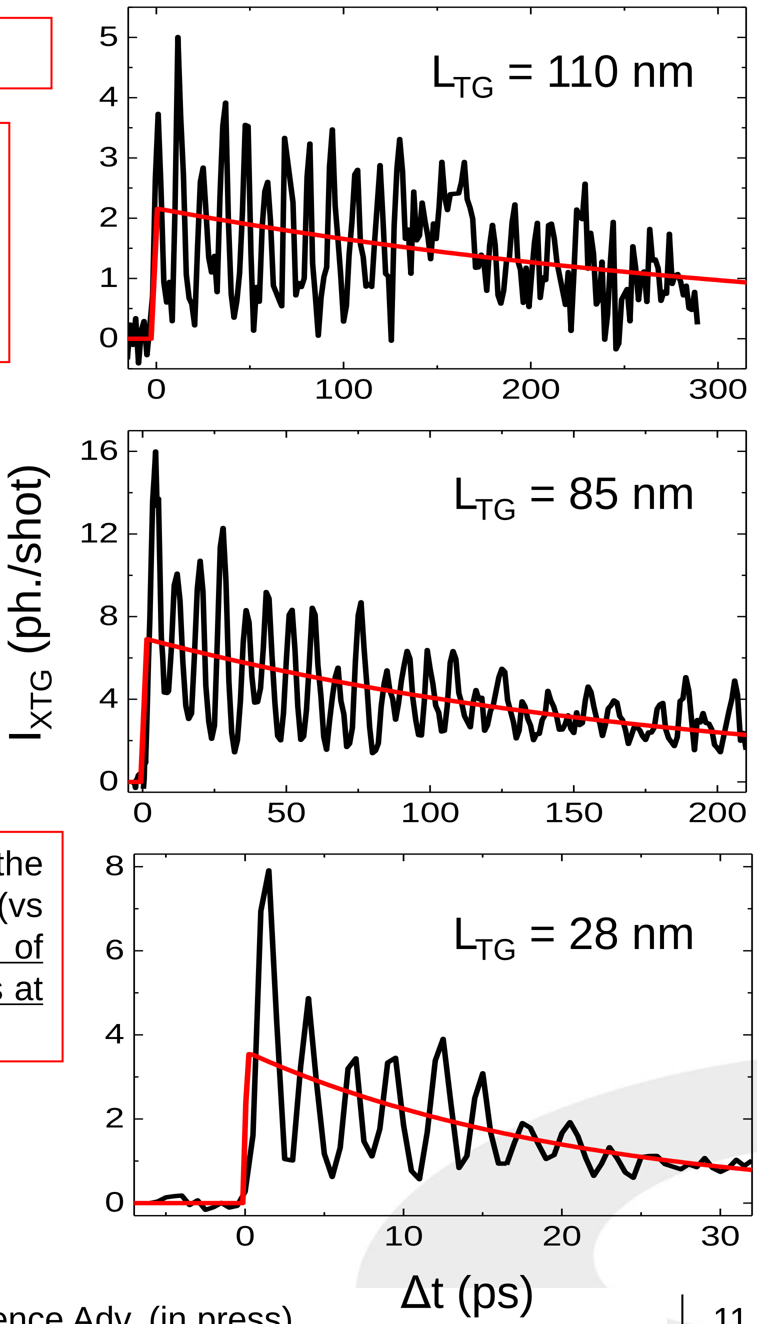
Sample: amorphous-Si_3N_4



L_{TG} scan at fixed crossing angle (θ) and varying λ_{FEL} (54 – 13 nm)

(red lines are the thermal decay)

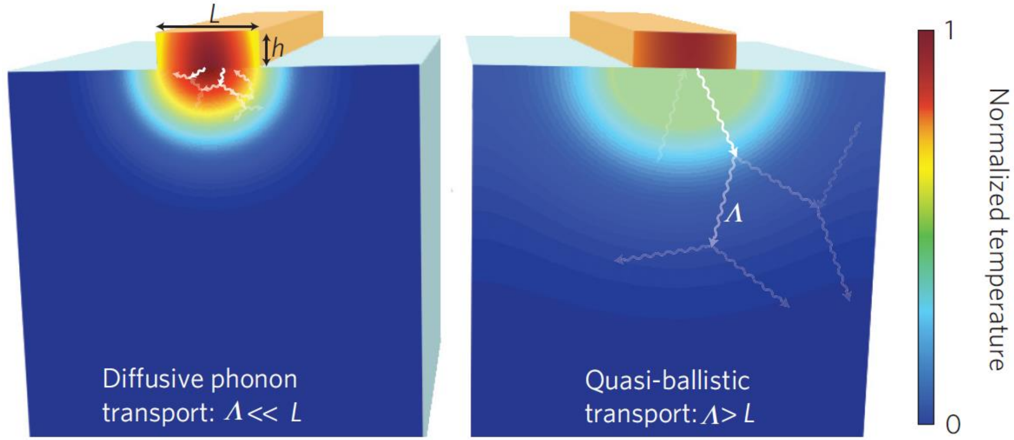
The signal modulation frequency and the thermal decay rate scale as expected (vs L_{TG}) → stimulated (Brillouin) scattering of EUV light and heat transport processes at 10's nm scales.



F. Bencivenga et al., Science Adv. (in press)

Nanoscale thermal transport

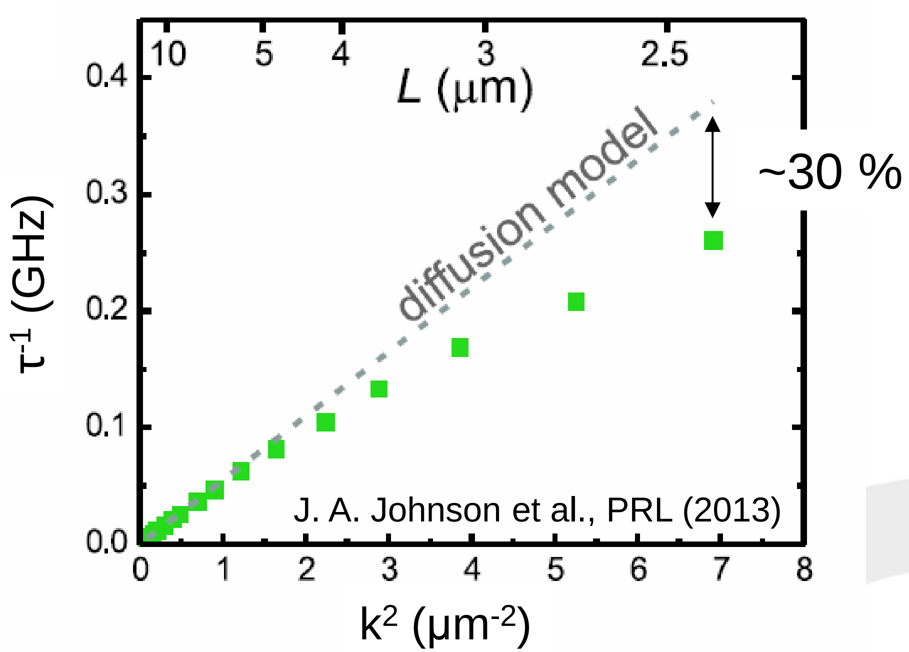
Heat transport at the nanoscale is a hot area of research[1]
→ thermal management of nanoelectronic devices, thermoelectric energy conversion, etc.



M. E Siemens et al. Nat. Materials (2010)

When the characteristic dimensions of the thermal gradient starts to compare with the phonon mean free path the Fourier law of diffusion breaks down. So, the question is:
<u>how fast is thermal relaxation process at the nanoscale?</u>

Optical TG: tangible deviation from the Fourier law (diffusion model) in Silicon on a few µm scale[2]



Nanoscale thermal transport

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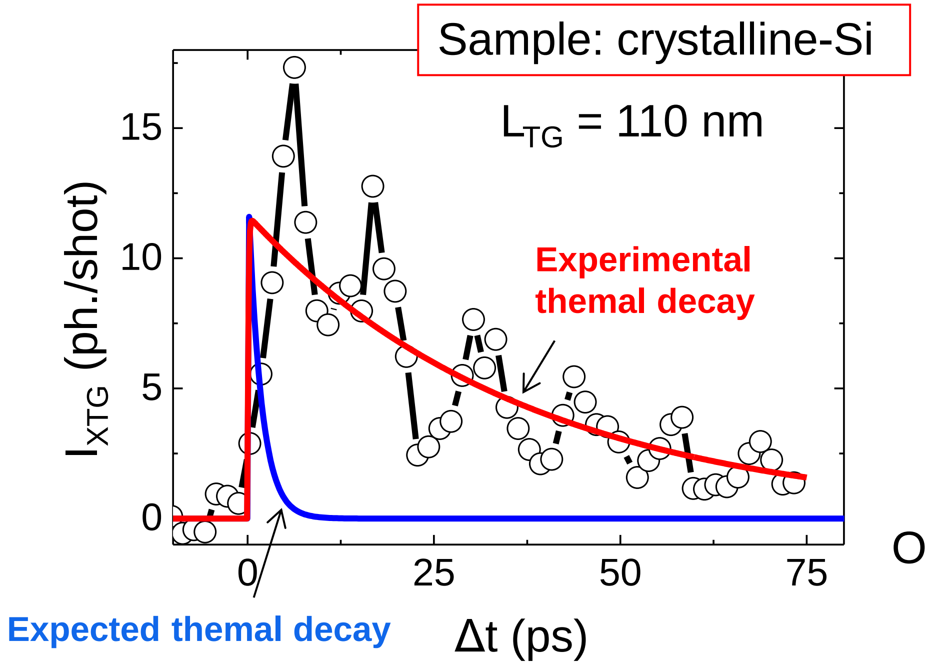

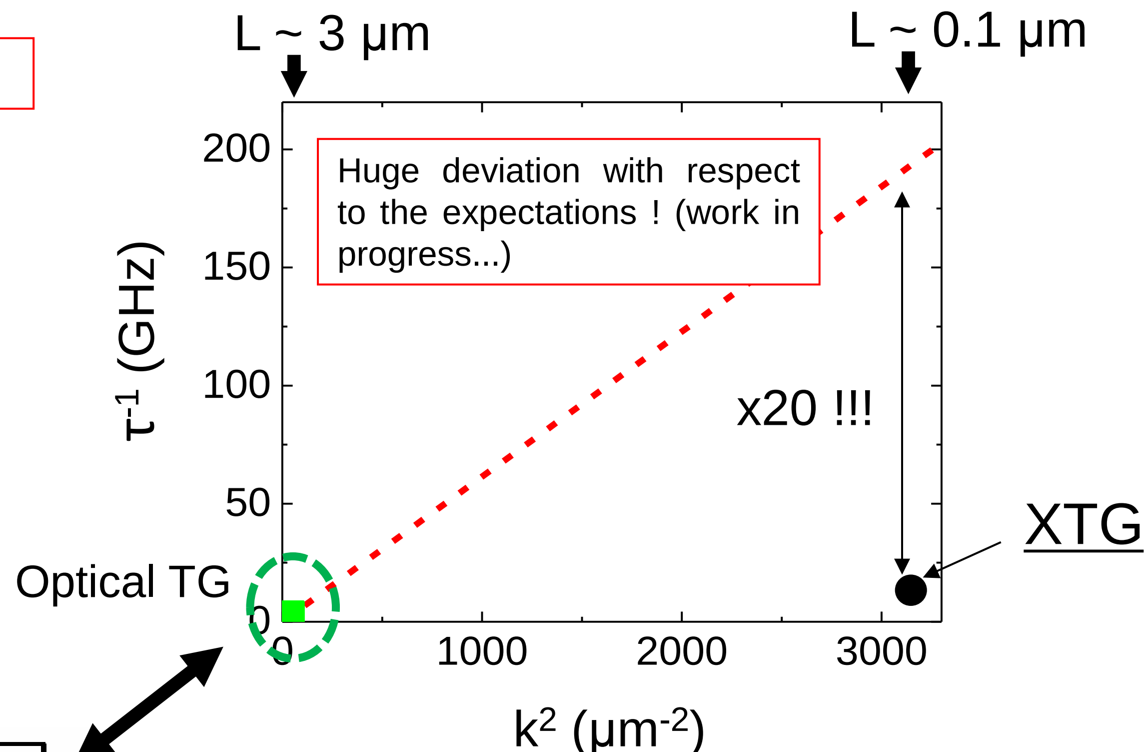

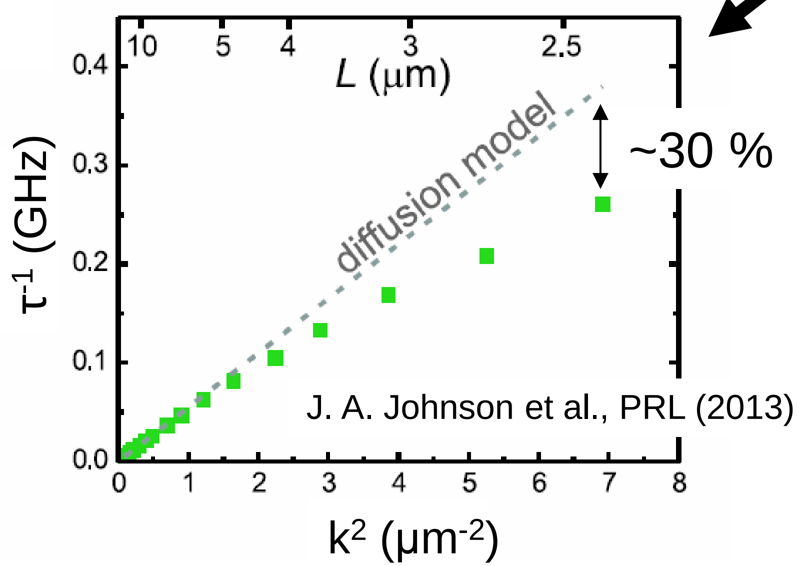

Collaboration (phonons and thermal dynamics):
G. Chen, K. A. Nelson, A. A. Maznev et al. (MIT); H. Capteyn, M. Murmane et al. (JILA); G. Monaco et al. (Uni. Trento); F. Bencivenga, F. Capotondi, C. Masciovecchio et al. (Elettra)

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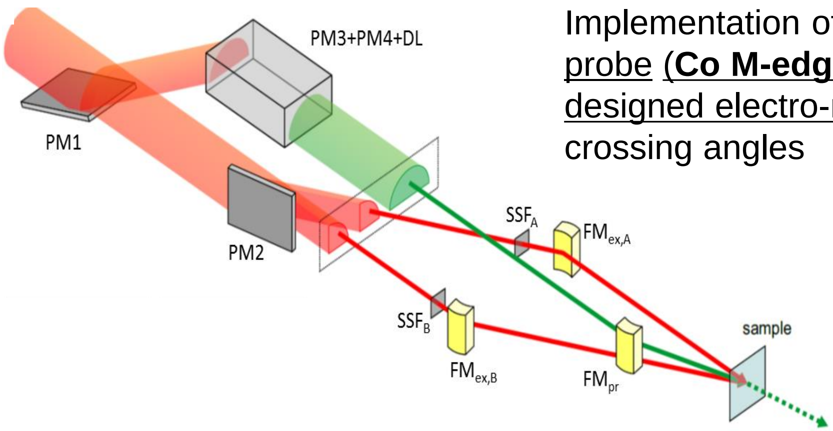
F. Bencivenga et al., Science Adv. (in press)

Beyond phonon/thermal dynamics

Nanoscale magnetic dynamics

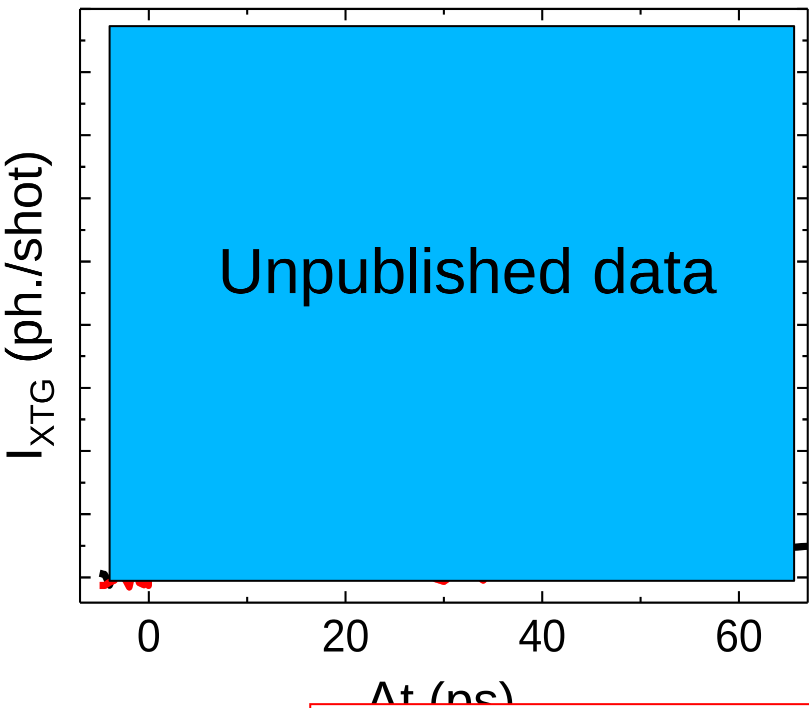


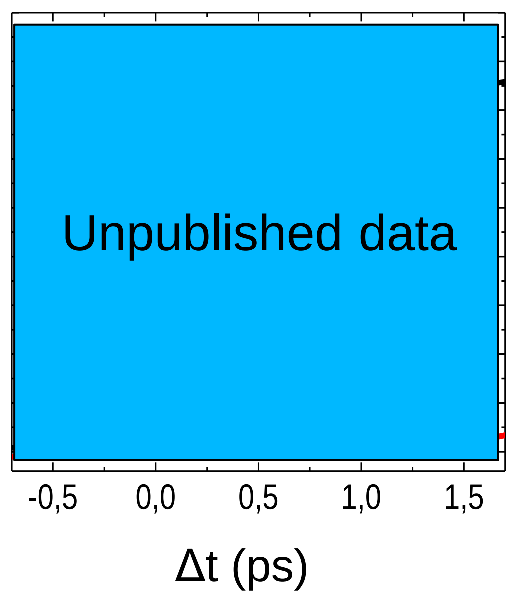
Implementation of the setup with <u>resonant probe</u> (**<u>Co M-edge</u>**; 60 eV) and a <u>specially designed electro-magnet</u> for XTG at wide crossing angles

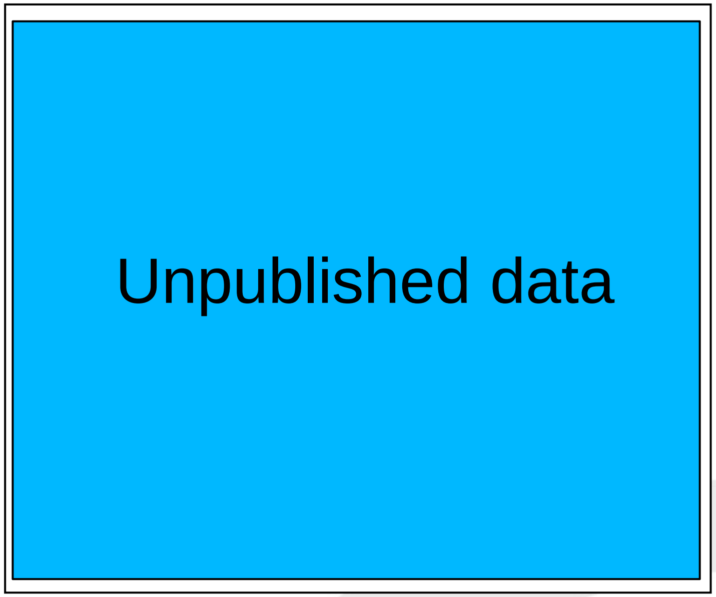
Collaboration: **<u>C. Gutt</u>**, D. Ksenzov, L. Randolph, H. Rahamann (Uni. Siegen); K. A. Nelson, **<u>A. A. Maznev</u>** (MIT); **<u>S. Bonetti</u>**, M. Pancaldi, V. Unikandanunni (Uni. Stockholm); S. Urazhdin (Emory Univ)

UNIVERSITÄT SIEGEN

Stockholm University







EUV TG may become a tool to probe magnetic, electronic and thermoelastic dynamics in a single experiment, with ultrafast time resolution and on the 1-100 nm length-scale

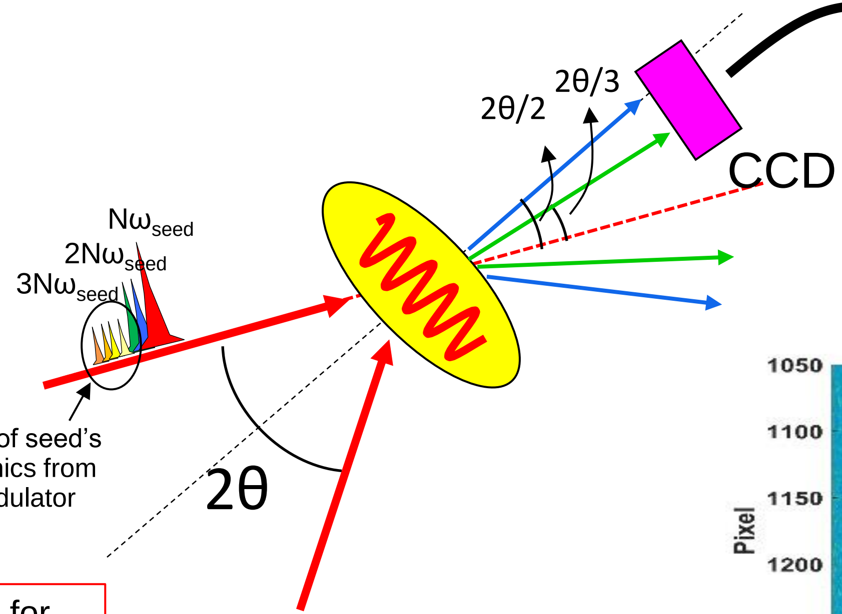
Beyond phonon/thermal dynamics
Four-wave-mixing from multi-eV excitations

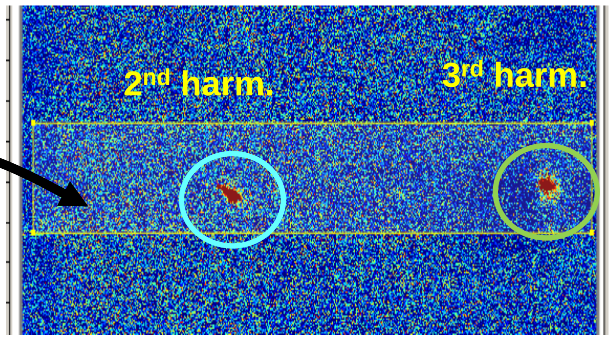
(from the intro): EUV TG is a 3rd order non-linear process and can be regarded as a starting point for developing XUV/soft x-ray four-wave-mixing (FWM) experiments[1]
The FERMI FEL can operate in multi-harmonic mode ($\omega_{FEL,N} \sim N\omega_{seed}$; $\omega_{seed} \sim$ 3-5 eV and N~5-50) with photon-energy and polarization tunability (but with some limitations)

1) S. Tanaka and S. Mukamel, PRL 2002 (and many other works)

First step: e a genuine time-zero EUV-FWM signal using as a probe the 'natural' FEL harmonics (i.e. $2N\omega_{seed}$ and $3N\omega_{seed}$,) which propagate along with the fundamental[2]

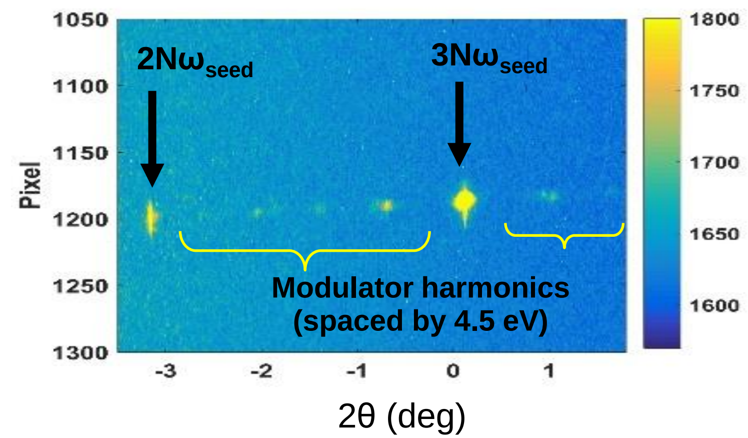
2) L. Foglia et al., PRL (2018)





Second step: optimize the setup for observing very small signals (e.g. the harmonics from the FEL modulator)[3]

3) R. Mincigrucci et al., NIMA (2018)



Try a real experiment (to be done!) looking at the multi-eV shifted FWM emission from the sample, meaning:
→ Frequency resolved FWM, by scanning the input frequencies and/or spectrally resolving the FWM signal.
→ Phase resolved FWM, by controlling the relative phases of FEL harmonics [K.C. Prince et al., Nat. Phot. (2018)]

Conclusions

1) FEL-based four-wave-mixing experiments in a transient grating scheme have been demonstrated at the FERMI seeded FEL and are now exploitable by users.
2) The XTG-pump/FEL-probe setup has allowed to reach spatial periodicities down to 24 nm ($|\mathbf{k}|$~0.26 nm^{-1}), XTG periodicities down to ~12 nm are straightforward using the present setup, the single-digit nm regime ($|\mathbf{k}|$~1 nm^{-1}) is harder but possible.
3) In amorphous Si_3N_4 the thermal transport time follows the quadratic trend predicted by Fourier law of diffusion down to 28 nm (thermal length-scale ~9 nm), in crystalline Si the deviation from the classical regime is huge already at 110 nm (thermal length-scale ~35 nm). That's about the expected siatuation... quantitative analysis ongoing
4) XTG with resonant probe (@Co M-edge) shows a sizable magnetic and electronic signal. XTG may become a nice tool for ultrafast magnetic studies at the nanoscale.
5) Four-wave-mixing (time-zero) signals stimulated by multi-colour FEL pulses have been observed and may be explited in frequency/phase resolved experiments.

Many thanks to...

Experimental team (TIMER and DiProI beamlines)

F. Capotondi
L. Foglia
M. kiskinova
C. Masciovecchio
R. Mincigrucci
E. Pedersoli

and former members...
A. Battistoni
R. Cucini
D. Naumenko
I. Quints-Lopez
A. Simoncig

Laser team (M. Danailov)
Machine physics team (L. Giannessi)
PADReS team (M. Zangrando)
FERMI menagement (M. Svandrlik)

... and all the FERMI team!

Collaborators (our first users!)

K. A. Nelson & A. A. Maznev (MIT)
G. Monaco (Univ. Trento)
C. Gutt (Siegen University)
S. Bonetti (Stockholm and Venice Univ.)